TOTAL- AND METHYL-MERCURY RESPONSE IN BRINE AND BIOTA TO DESTRATIFICATION OF THE GREAT SALT LAKE, UTAH, USA

by

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STATEMENT OF THESIS APPROVAL

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ABSTRACT

Measurements of the aquatic chemistry of the South Arm of Great Salt Lake were made prior to and after elimination of flow between its North and South Arms to demonstrate dramatic alteration of the system in response to sealing of the railway causeway separating the arms. Perennial stratification of oxic shallow and anoxic deep brine layers characteristic of the South Arm prior to sealing of the causeway has been replaced by a vertically homogenous oxic brine following elimination of flow between the arms. Total mercury concentration in the water column and underlying sediment decreased to approximately 20% of their pre-causeway sealing values, and methyl mercury concentrations became insignificant relative to pre-sealing values. These measurements provide new understanding of the role of the deep brine layer in promoting the highly elevated methyl mercury concentrations observed before causeway sealing. Additional mercury measurements in biota provide further information regarding the implied connection between elevated methyl mercury concentrations in the deep brine layer and elevated mercury in avian species reported prior to causeway sealing.

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CHAPTER 1

INTRODUCTION

Mercury is a global pollutant that spreads through natural systems by complex transformation and transport processes that are in large part governed by redox conditions in the environment (Hsu-Kim et al., 2013). Mercury exists in nature as elemental mercury Hg(0), divalent mercury Hg(II), and organomercury compounds such as monomethylmercury (MeHg), the bioaccumulative form of mercury. Biomagnification of MeHg can result in potentially toxic levels of MeHg exposure to humans and wildlife through consumption of prey, causing detrimental neurological, behavioral, and reproductive effects (Mergler et al., 2007).

The methylation of inorganic mercury in the environment is predominantly biologically mediated, occurs in anoxic environments, and is carried out largely by sulfate-reducing bacteria (King et al., 2000; Sunderland et al., 2006; Graham et al., 2012). Methylation by iron-reducing bacteria (Fleming et al., 2006; Kerin et al., 2006) and methanogenic bacteria (Hamelin et al., 2011) can also be important, and many other microbes have been shown to contain the *hgcAB* gene cluster responsible for mercury methylation (Gilmour et al., 2013). DOM has been shown to increase mercury methylation under sulfidic conditions in lab experiments (Graham et al., 2012, 2013) and MeHg concentrations in aquatic sediments are often positively correlated with

concentrations of dissolved organic carbon (DOC) (Marvin-DiPasquale et al., 2009), although this is not always the case (Hammerschmidt et al., 2008) due to the complex effects of DOM on mercury's biogeochemical cycling via its role as a substrate for microbial respiration and its role in inorganic mercury complexation and chemical speciation (Benoit et al., 2003)

The GSL is the largest terminal lake in the Western Hemisphere, and the Western Hemisphere Shorebird Reserve Network recognizes it as a habitat of hemispheric importance for millions of migratory birds (Figure 1). Over 1.4 million shorebirds use the GSL for breeding and staging areas, and over 7 million waterfowl utilize the GSL and adjacent 1,900 km^2 of freshwater and brackish wetlands during some portion of their biannual migration. The GSL avian community feeds on aquatic organisms adapted to highly saline conditions that include brine shrimp (*Artemia fransicana*), brine fly (*Ephydra spp.*), water boatman (*corixid*), and numerous algal species (Loving et al. 2002). In 2007, human consumption advisories were placed on three duck species (Cinnamon Teal, Northern Shoveler, and Common Goldeneye) at the Great Salt Lake based on mercury levels in breast muscle tissue exceeding the EPA screening value (0.3 µg/kg, ww) (Scholl and Ball, 2005 and 2006).

The construction of a railroad causeway in 1959 restricted water flow between the North and South Arms of the lake (Gwynn, 2002). Because the South arm receives nearly all freshwater inputs to GSL, the North Arm was evaporatively concentrated to a greater extent than the South Arm, yielding brine in the North Arm (250-280 g/L) that is 1.4 to 1.6 times more saline than the South Arm (110-180 g/L), the latter being 3–5 times more saline than seawater (33-36 g/L) (Arnow and Stephens, 1990). Openings in the

causeway allowed limited density-driven bi-directional flow between the North and South Arms, resulting in stratification and the formation of a deep brine layer (DBL) in the South Arm (Loving et al., 2002) that persisted between 6.5 and 9 m below the surface of the lake, and that was not subject to annual turnover (Naftz et al., 2008; Diaz et al., 2009). The DBL was anoxic with elevated DOC (59 – 88 mg/L) and sulfide (7 –29 mg/L) (Gwynn, 2002; Diaz et al., 2009; Johnson et al., 2015), high activities of sulfate-reducing bacteria (Ingvorsen and Brandt, 2002), and elevated MeHg (20 to 32 ng/L) and total mercury (38-80 ng/L) with a high percentage of the HgT existing as MeHg. The elevated MeHg in the DBL implicates it as a potential source of the elevated mercury in biota in surrounding areas (Naftz et al., 2008; Vest et al., 2009; Wurtzbaugh et al., 2011; Saxton et al., 2014).

Although there was persistent stratification in the GSL, Beisner et al. (2009) provide evidence that limited mixing between the DBL and shallow brine layer occurred during conveyance of the DBL from the northern to southern areas of the South Arm, indicating the potential transfer of MeHg from the DBL to overlying shallow brine layer, and by extension, to brine fly larvae and brine shrimp that serve as diet for many avian species (Belovsky et al, 2011; Roberts, 2013; Roberts and Conover, 2014). Dissolved inorganic nitrogen (DIN) and total dissolved phosphorous (TDP) in the South Arm of the GSL reflect two distinct nutrient pools in the Upper Brine Layer and the Deep Brine Layer, respectively (Belovsky et al., 2011).

Spatial and temporal trends further suggest that the DBL may be a predominant source of the MeHg found in the surface ecosystem. For example, methylmercury production potentials (MPP) in the DBL were higher in spring (April) relative to summer

(July) (Johnson et al., 2015); similarly, concentrations of HgT in brine shrimp and brine flies are also lower during mid-summer relative to other times of year (Peterson and Gustin, 2008; Van Leeuwen et al., 2011; Wurtsbaugh et al., 2011). Furthermore, blood HgT concentrations in Eared Grebes (Vest et al., 2009) and HgT concentrations in brine flies (Wurtsbaugh et al., 2011) were greater at locations where the South Arm was the closest major water body. Lower HgT concentrations in Eared Grebes and brine flies were measured in areas proximal to freshwater-influenced bays despite the sediment and water being elevated in MeHg, which includes areas near Farmington Bay and sheet flow wetlands with elevated MeHg concentrations in sediment and surface waters (Johnson et al., 2015). Although the above temporal and spatial trends are suggestive, no direct link has been demonstrated between the elevated concentrations of MeHg in the DBL and the elevated concentrations of mercury in waterfowl or other biota at the GSL.

In late 2012, one of the two railway causeway culverts that allowed limited water flow between the North and South Arms of the GSL was closed due to deteriorating structural integrity. The second culvert was closed in December 2013, sealing the causeway to flow. This event provided the opportunity to determine the geochemical response of the system to elimination of flow between the North and South Arms and the resulting destratification and disappearance of the DBL in the South Arm (described below). This event also allowed us to quantify how HgT and MeHg concentrations in the water column and biota responded to the disappearance of the DBL, thus providing insight into the role of the DBL on the biogeochemical cycling of mercury in the GSL.

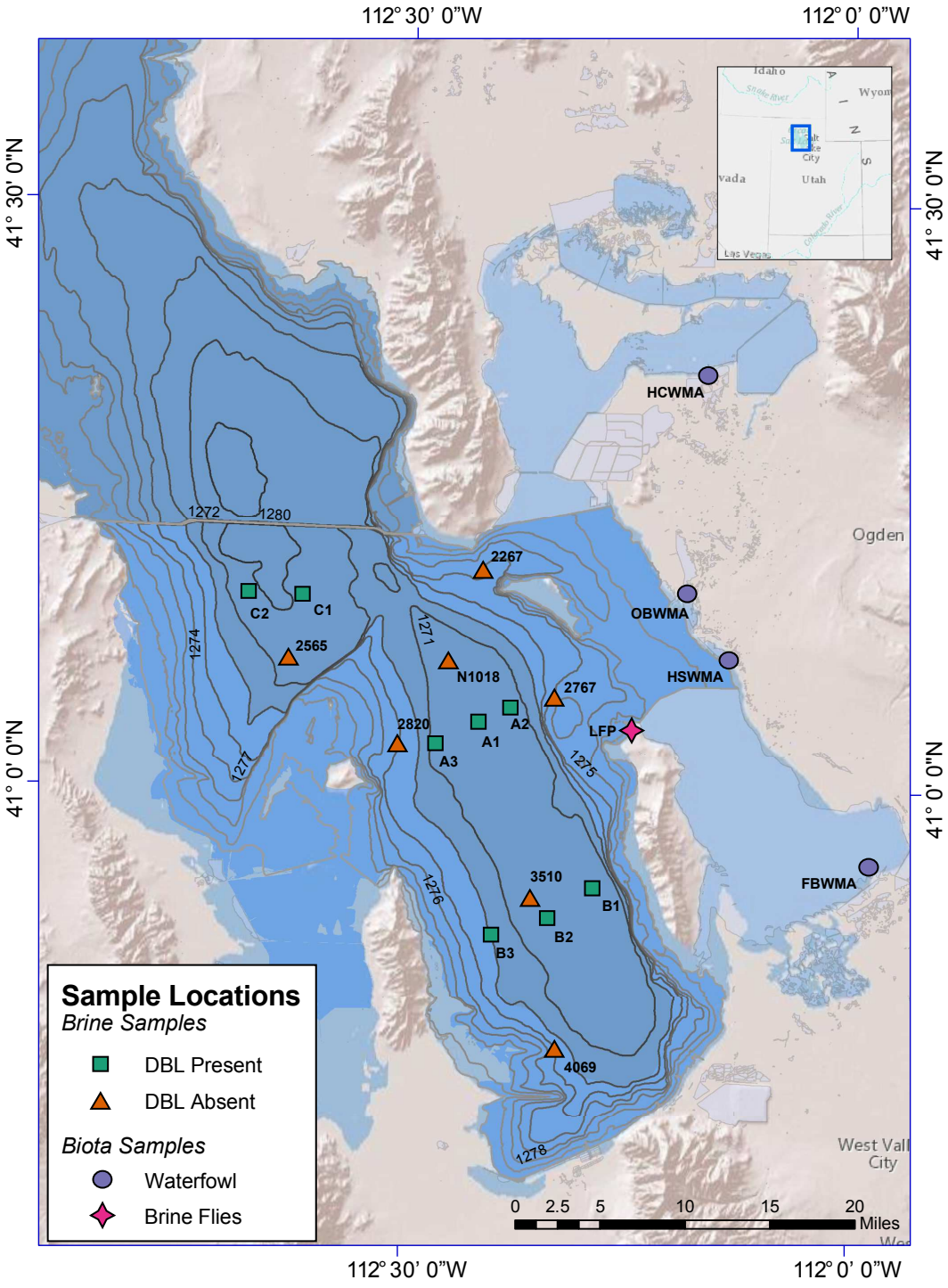

Figure 1. Locations sampled for brine and biota samples. Shading represents typical salinities (% total dissolved solids) observed prior to closure of the culverts, with gray corresponding to approximately 28%, darker blue corresponding to approximately 15%, and the light blue representing salinities in the range from 5% to 0.5%. Thus, the darker shading in the South Arm also indicates the approximate extent of the deep brine layer prior to closure of the culverts.

CHAPTER 2

METHODS

2.1 Water and Sediment Sampling and Analysis

In October 2015, water column chemical and physical conditions were characterized at five locations across the South Arm of GSL and Farmington Bay (Figure 1). Water samples were collected at five stations (2267, 2565, 2767, N1018, and 3510) at 0.2 m below lake surface and 0.5 m above lake bottom, referred to as shallow and deep samples, respectively. The data from these samples collected after the disappearance of the DBL were compared to previous data collected from the shallow and deep brine layers from May 2009 to December 2012, prior to causeway closure when the DBL was present (Johnson et al., 2015) (Figure 1). Water column temperature, specific conductance, pH, and dissolved oxygen (DO) were measured in the field using a YSI Professional Series Quatro probe. The probe was calibrated within 12 hours prior to sampling. Sulfide was measured in filtered water column samples in the field immediately after collection using a photometric method (V-2000 Multi-analyte LED Photometer and Vacu-vials®, CHEMetrics).

Clean Hands -- Dirty Hands protocol (US EPA, 1996) was followed during Hg sample collection and analysis. Unfiltered Hg water samples were collected by peristaltic pump using acid-washed PTFE tubing into precleaned and double bagged triple-rinsed

FLPE bottles (precleaned by triple rinsing with Milli-Q water and drying in a laminar flow hood). Bottles used to collect anoxic water samples were filled to overflowing in order to minimize headspace. Filtered water samples were passed through 0.45-micron pore size, pre-acid rinsed capsule filters in the field (Geotech Environmental). After collection, water samples for total mercury (HgT) and methylmercury (MeHg) analyses were stored on ice in the field and acidified to 0.5% using trace-metal grade sulfuric acid for preservation the same day as sampling. All samples were then refrigerated and analyzed within two weeks of collection

Total mercury (HgT) samples were oxidized by amendment to 5% BrCl at least 24 hours prior to analysis. This higher BrCl concentration relative to established protocols (US EPA, 2002) that call for 1% BrCl was necessary to fully oxidize the high levels of dissolved organic matter (75.4 ± 11.1 mg/L) in these water samples. Total Hg concentrations in water were determined via reduction with $SnCl_2$, purge and trap onto gold traps, and subsequent thermal desorption with quantification by cold vapor atomic fluorescence spectroscopy (CVAFS) using a MERX-T automated system (Brooks Rand) using established techniques (US EPA, 2002). Recoveries of HgT in the USGS Standard Reference Sample Hg-43 for HgT in water averaged 75 to 125%. MeHg concentration in water was measured after distillation with ammonium pyrrolidine dithiocarbamate (APDC), followed by aqueous phase ethylation, purge and trap onto tenax traps, thermal desorption, pyrolitic decomposition, and CVAFS detection using a MERX-M automated system (Brooks Rand using established techniques (US EPA, 2001). Because no certified reference material exists for MeHg in water, MeHg matrix spikes were included in each distillation, MeHg spike recoveries averaging 75 to 125%. Water samples for dissolved

organic carbon (DOC) analysis were placed on ice in the field then frozen in the lab each evening. DOC was measured in water samples (TOC-5000a, Shimadzu, Inc.) within one week of sample collection using EPA Method 1684 (2001).

Surficial sediment from the GSL sampled (Figure 1) was collected by peristaltic pump using acid-washed PTFE tubing into precleaned and double bagged triple-rinsed 500 mL FLPE bottles (precleaned by triple rinsing with Milli-Q water and drying in a laminar flow hood) filled to overflowing to minimize headspace, and stored on ice in the field. THg extraction from sediment samples involved digestion in a 7:3 TMG HNO3/H2SO4 solution (80C° for 6 hours) and dilution in 5% BrCl (EPA Method 1631-appendix). For MeHg extraction, 1 g of sediment was reacted with 5 mL of KBr/H2SO4 solution (18% w-v-1 KBr, 5% v-v-1 TMG H2SO4) and 1 mL CuSO4 (1 M), and subsequently extracted with methylene chloride (10 mL). An aliquot of the extraction solvent (2 mL) was pipetted into a Teflon distillation tube with 50 mL of MilliQ water, followed by complete evaporation of methylene chloride (1- 2 hours at 55°C, purged with ultra-high purity N2) (Bloom et al.).

2.2 Biota Collection and Analyses

Adult brine flies were collected from Lady Finger Point on Antelope Island of the GSL (Figure 1) from spring to late fall of 2012 to 2015. Flies were collected with nets, transferred directly placed into polypropelene tubes, placed on ice in the field, and frozen back in the lab the same day. Seventy-six waterfowl were harvested in mid-September of 2014 and ninety-nine were harvested in late August and early September of 2015 from the Great Salt Lake and surrounding wetlands by the Utah Division of Wildlife Resources

in order to compare the change in mercury burden before and after the disappearance of the DBL. The waterfowl were harvested from the Farmington Bay, Ogden Bay, Howard Slough, and Harold Crane Waterfowl Management Areas (Figure 1). Decoys were used to attract ducks to the location of collection and all were shot with shotguns using non-lead shot. Upon collection, specimens were placed in ziplock bags and frozen. The age of each bird was determined by looking at physical characteristics of the birds, examining rectrices, wing and body plumage, and cloacal characters. The species sampled were Northern Shovelers (*Anas clypeata*) (DBL Present: n = 16; DBL Absent: n= 16), Mallard (*Anas platyrhynchos*) (DBL Present: n= 16; DBL Absent: n= 21), Gadwall (*Anas strepera)* (DBL Present: n = 15; DBL Absent: n =22), and Cinnamon Teal (*Anas cyanoptera)* (DBL Present: n = 23; DBL Absent: n = 31). Tissue sample collection involved thawing the birds for 12 hours and using a scalpel to cut skin away from muscle tissue. Five grams of breast muscle tissue per duck was harvested and stored in a Whirl-Pak (NASCO) polyethylene bag and frozen. Breast muscle was analyzed because it is the most likely muscle group to be consumed by duck hunters.

Both brine fly and waterfowl samples were freeze-dried and homogenized prior to analysis, and thus all HgT concentrations in biota are reported on a dry weight basis. Brine flies were digested in Teflon vials with 10 mL of a 2:1 mixture of trace metal grade nitric and sulfuric acids. Samples were allowed to predigest at room temperature for 1 hour, then heated to 100 °C for 4 hours. Following the digestion, samples were amended to 1% BrCl and the sample diluted to 50 mL with ultra-high purity water. All digestions included at least 2 digestion blanks and 2 certified reference materials (TORT-2 and DORM-3, National Research Council Canada), each digested in duplicate. Aliquots of

the digested samples were measured by oxidation with BrCl, neutralization with hydroxylamine hydrochloride, reduction with $SnCl_2$, purge and trap using dual-stage gold trap amalgamation, and quantification by CVAFS (US EPA, 2002). The average daily HgT detection limit, based on 3 times the standard deviation of digestion blanks, was 0.007 ng g^{-1}, assuming a 100 mg sample. Recoveries for HgT in the biota certified reference materials averaged 101.4% +/- 7.8% (n = 88). The duck muscle tissue was analyzed using thermal decomposition, amalgamation, and atomic absorption spectrophotometry using a DMA-80 (Milestone) following established protocols (US EPA, 2007). Muscle tissue samples were only analyzed for HgT because previous studies have shown that >95% of Hg in bird muscle is MeHg (Evers et al., 2005). Each analysis run included each of the two certified reference materials (TORT-2 and DORM-3) analyzed in duplicate, with recoveries of HgT ranging from 88% to 123%. Analytical precision of the HgT measurements was assessed by analyzing eight samples in triplicate. The average percent relative standard deviation of these triplicate measurements was 5.2%.

The HgT concentrations in the waterfowl were log transformed prior to statistical analysis in order to meet the assumptions of parametric statistics. The data were analyzed using multifactor ANOVA using a crossed design. While we did not anticipate *a priori* that the sex of the waterfowl would have any effect on their HgT concentrations, we initially included sex in the statistical model, which included the following five variables as fixed factors: duck species, age, year, site, and sex. The fully crossed model could not be run due to missing cells because not all sites were sampled both years. We therefore used a reduced model (Winer et al., 1991) made by removing the five-way and

four-way interactions. In this model, none of the interactions involving sex nor sex as a main factor were even remotely significant ($p>0.49$ in all cases). Given this result along with our *a priori* assumption regarding the importance of sex, this factor was removed from the model. The multifactor ANOVA using a crossed design with the fixed factors duck species, age, year, and site was then reduced (Winer et al., 1991) by removing the four-way interaction and the three-way interaction Age × Site × Year due to the missing cells, as described above.

CHAPTER 3

RESULTS

3.1 Aquatic Field Parameters

In 2008 to 2012, prior to closure of the causeway culverts, shallow waters in the South Arm were at or near DO saturation (9.48 ± 0.70 mg/L), whereas deep waters in areas where the DBL exists were anoxic (DO = 0.05 ± 0.02) (Figure 2). After causeway closure, mean DO was 9.1 ± 3.4 mg/L in shallow waters and 7.2 ± 5.2 mg/L in all deep waters. Prior to culvert closure, DO concentrations were significantly lower in surface waters than in the DBL (two-tailed t-test, unequal variance, $p << 0.001$), whereas there was no significant difference in DO between shallow and deep waters ($p = 0.52$) two years after the culverts were closed. Under pre-closure conditions (before 2012), mean pH in shallow (8.35 ± 0.71) and deep waters (7.62 ± 0.12) differed significantly (t-test, $p = 0.01$). However, two years after the culvert closures, mean pH was 8.27 ± 0.40 and 8.09 ± 0.20 for shallow and deep waters, respectively, with no significant difference ($p = 0.39$) (Figure 2). Under pre-closure conditions, mean sulfide concentrations in filtered shallow waters (1.3 ± 1.0 mg/L) were significantly lower ($p = 0.001$) than sulfide concentrations in filtered deep waters (19.2 ± 7.6 mg/L). After culvert closure, there was no longer a difference ($p = 0.51$) between mean sulfide concentrations in filtered shallow waters (0.04 ± 0.02 mg/L) and deep waters (0.05 ± 0.01 mg/L) (Figure 2). Before

causeway closure, mean DOC concentrations in the shallow waters was 49 ± 12 mg/L compared to 75 ± 11 mg/L in the pre-closure DBL, with these being significantly different ($p = 0.005$). After causeway closure, mean DOC concentration in shallow waters (61.9 ± 1.2 mg/L) and deep waters (62.4 ±2.2 mg/L) were no longer significantly different ($p= 0.75$) (Figure 2). Profiles developed using data from the USGS and the State of Utah for DO, temperature, and specific conductance (SC) demonstrate the loss of the sharp interface between the deep and shallow brines after causeway closure (Supporting Information).

3.2 Mercury Concentrations in Water and Surficial Sediment

Prior to causeway closure, mean HgT concentrations were 54.7 ng/L ± 0.47 and 2.81 ng/L ± 0.05 in the deep and shallow brines, respectively (Figure 3), yielding a significant difference ($p << 0.001$) for HgT. Mean MeHg was 25.46 ± 4.24 ng/L and 0.49 ± 0.05 ng/L in the deep and shallow brines, respectively, yielding a significant difference ($p << 0.001$) for MeHg. Following culvert closure, mean HgT concentration was 6.35 ± 2.46 ng/L and 5.16 ± 1.25 ng/L in deep and shallow samples, respectively, and mean MeHg concentration was 0.78 ± 0.54 ng/L and 0.64 ± ng/L, in deep and shallow samples, respectively. These results showed no significant difference between deep and shallow samples for HgT ($p=0.14$) or MeHg ($p= 0.59$). Prior to culvert closure, the fraction of HgT comprised of MeHg in deep brine was 30 to 80% (Johnson et al., 2015), whereas following causeway closure, this ratio decreased to 2.5 to 21%.

Average HgT and MeHg concentrations in surface sediment underlying the DBL in the South Arm (before culvert closure) were 103 ± 80 µg/g (d.w.) and 0.86 ± 0.54

µg/g (d.w.), respectively (Figure 4). Average HgT and MeHg concentrations in surface sediment following causeway sealing were 27 ± 17 µg/g (d.w.) and 0.18 ±0.08 ppm (d.w.), respectively. Thus, HgT concentrations in surface sediment decreased by roughly 75% ($p = 0.069$) in response to causeway closure, whereas MeHg concentrations in surface sediments decreased by roughly 80% ($p = 0.025$) during this same time period.

3.3 Mercury Concentrations in Biota

Mean HgT concentrations in adult brine flies were 446.5 ± 128 ng/g (dw) and 534 ± 204 ng/g (dw) for DBL-present versus DBL-absent conditions, respectively, with this decrease being statistically significant ($p << 0.001$). Brine fly Hg burdens separated by month (Figure 5) show that the higher HgT concentrations under DBL-absent conditions are associated with summer months.

Mean HgT concentrations in all waterfowl were 440 ± 480 ppb (dw) for pre-closure versus 410 ± 380 ppb (dw) for post-closure conditions, respectively. Waterfowl Hg concentrations according to species (Figure 6) show mean HgT concentrations in pre-closure versus post-closure samples were 480. ± 230 ppb (dw) versus 690 ± 480 ppb (dw) (Cinnamon Teal), 67 ± 49 ppb (dw) versus 140 ± 220 ppb (dw) (Gadwall), 130 ± 160 ppb (dw) versus 230 ± 270 ppb (dw) (Mallard), and 920 ± 300 ppb (dw) versus 640 ± 590 ppb (dw) (Northern Shovelers).

There was a significant effect ($p << 0.001$) of duck species on their HgT concentration according to multifactor ANOVA analysis using a crossed experimental design including the fixed factors: duck species, age, site, and year (Table 1). Northern shovelers and cinnamon teal had the highest HgT concentrations, followed by mallards

and gadwalls with the lowest HgT concentrations. Tukey pair-wise post-hoc comparisons demonstrated that HgT concentrations in northern shovelers and cinnamon teal were not significantly different ($p = 0.99$) while HgT in both mallards and gadwalls were significantly different from all other duck species ($p < 0.013$ in all cases). There was no significant difference ($p = 0.38$) pre- versus post-closure by age (Figure 7). Similarly, there was no significant difference in HgT concentrations in ducks by site ($p = 0.07$) (Figure 8).

This model was run after the removal of the four-way interaction and the three-way interaction Age × Site × Year due to incomplete data. The significant interaction ($p = 0.010$) between duck species and age (Figure 7) was due to the effect of age on HgT concentration not being the same in all duck species. Using planned contrasts, we found that there was a significant decrease in HgT from hatching year versus after hatching year for northern shovelers ($p = 0.011$) and mallards ($p = 0.016$), but there was no effect of age on HgT in cinnamon teal ($p = 0.48$) or gadwalls ($p = 0.59$) (Figure 6). This significant interaction ($p = 0.029$) between duck age and year (Figure 7) was due to a minor difference in HgT concentrations between 2014 and 2015 (corresponding to the two age classes). However, this interaction did not affect our ability to interpret the main effect (duck species versus HgT). A significant interaction ($p = 0.001$) was demonstrated between duck species and site (Table 1).

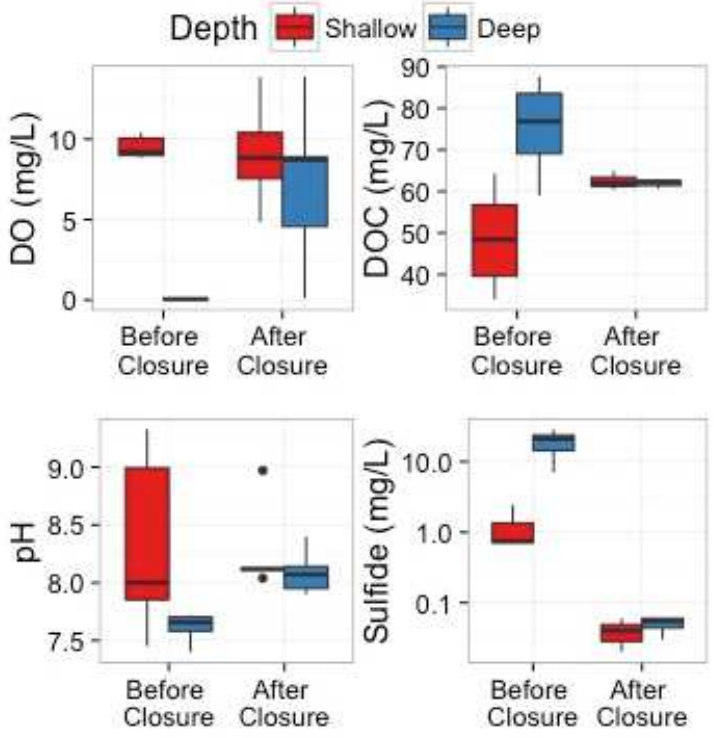

Figure 2. Dissolved oxygen (DO), pH, sulfide, and DOC concentrations in the shallow and deep brine layers with DBL present (before DBL disappearance) and after DBL disappearance. DBL-present: shallow (n= 9), deep (n=6); DBL-absent: shallow (n=5), deep (n=5)

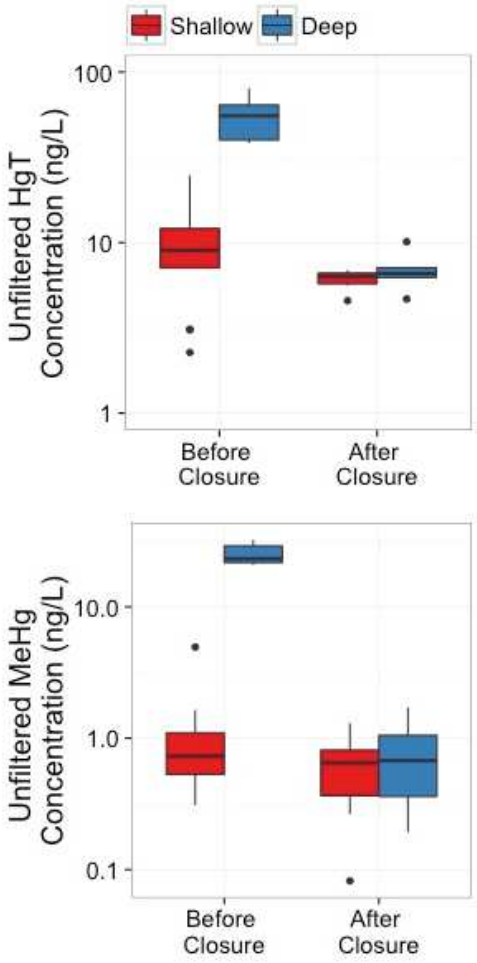

Figure 3: Unfiltered HgT and unfiltered MeHg concentrations from shallow and deep water (brine) samples from before (2008 to 2012) and after (October, 2015) the culvert closures in the GSL causeway. After DBL disappearance, unfiltered total mercury concentrations at depth ranged from 3.6 to 11.9 ng/L and unfiltered methylmercury concentrations ranged from 0.2 to 1.13 ng/L. DBL-present: shallow (n = 13), deep (n=9); DBL-absent: shallow (n=5), deep (n=5).

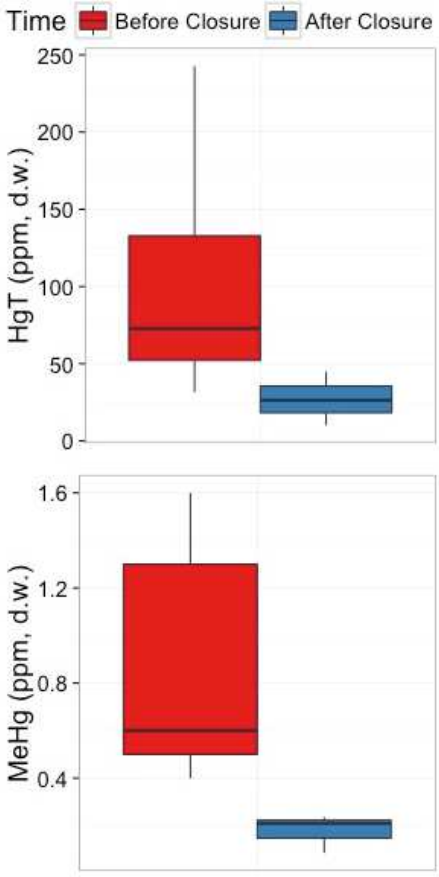

Figure 4: Blot plot of HgT and MeHg concentrations in surficial sediment collected from X sites underlying the DBL when the DBL was present (dates) and after its disappearance (October, 2015). Average HgT concentrations when the DBL was present were 103 ± 79.8 ppm (d.w.), and average MeHg concentrations were 0.86 ± 0.54 ppm (d.w.). After DBL retreat, average sediment HgT averaged 27.1 ± 17.3 ppm (d.w.) and sediment MeHg averaged 0.18 ±0.08 ppm (d.w.). There was a significant decrease in both HgT (p-value = 0.069) and MeHg (p-value= 0.025) following the disappearance of the DBL.

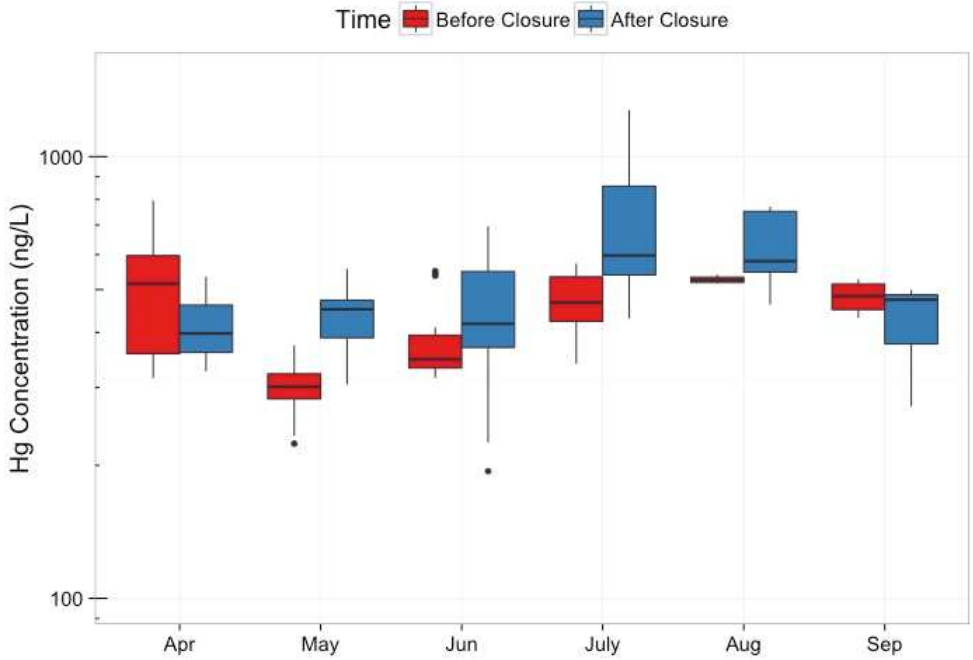

Figure 5: Concentrations of HgT in adult brine flies by month before culvert closure (2012-2014, n = 156) and after culvert closure (2015, n = 59) disappearance of the DBL.

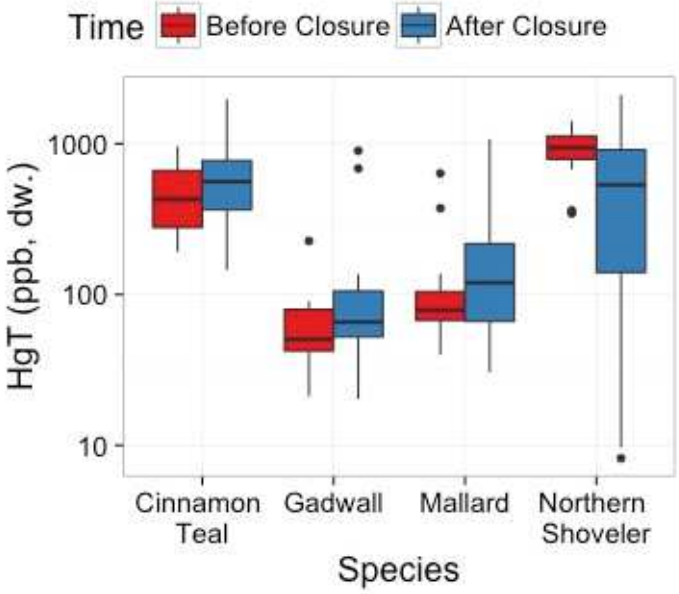

Figure 6. Total mercury concentrations in muscle tissue of waterfowl when the DBL was present (September, 2014) versus when absent (August-September, 2015).

Table 1. Multifactor ANOVA results for log transformed HgT in waterfowl muscle tissue using a crossed experimental design that included the four fixed factors duck species, age, site, and year. The model was run after the removal of the four-way interaction and one three-way interaction due to missing cells.

Source	Type III SS	df	Mean square	F-ratio	p value
Duck species	8.749	3	2.920	25.16	< 0.001
Age	0.798	1	0.798	6.89	0.010
Site	0.841	3	0.280	2.42	0.070
Year	0.092	1	0.920	0.79	0.375
Age × Site	0.351	3	0.117	1.01	0.392
Duck species × Age	1.378	3	0.459	3.96	0.010
Age × Year	0.567	1	0.567	4.89	0.029
Duck species × Site	3.257	8	0.407	3.51	0.001
Site × Year	0.055	2	0.280	0.24	0.788
Duck species × Year	0.878	3	0.293	2.53	0.061
Duck species × Age × Site	0.298	5	0.060	0.52	0.764
Duck Species × Age × Year	0.775	3	0.258	2.23	0.088
Species × Site × Year	0.579	2	0.289	2.50	0.086
Intercept	206.4	1	206.4	1780	< 0.001
Error	14.0	121	0.116		

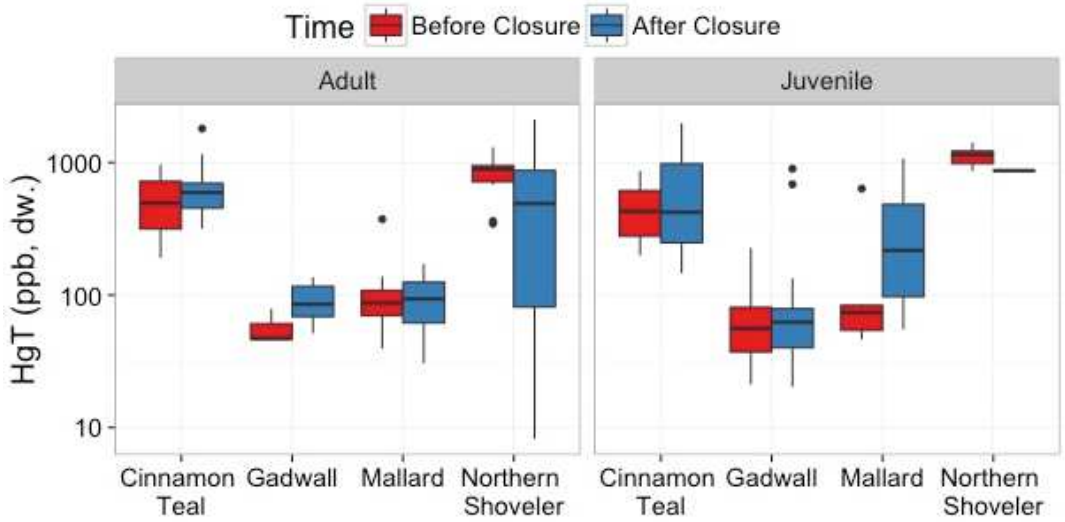

Figure 7: Mercury concentrations in juvenile (DBL Present: n =42; DBL Absent: n = 41) versus adult (DBL Present: n =28; DBL Absent: n = 49) waterfowl while the DBL is present compared to when the DBL is absent. There is no significant difference in mean mercury concentrations for adults or juvenile waterfowl.

CHAPTER 4

DISCUSSION

The GSL water column changed dramatically in response to closure of the railroad causeway. In response to causeway closure in 2014, the major contrast in DO, pH, and sulfide concentrations between shallow and deep samples were eliminated (Figure 2). The results demonstrate disappearance of the DBL in response to causeway closure such that post-closure, a single oxic brine occupies the water column from surface to bottom. Oxygen solubility decreases with increasing salinity, but vary widely across seasons with changes in water temperature and sunlight, which drive photosynthesis and oxygen super-saturation at peak activity (Carling et al., 2011).

The observed disappearance of measureable sulfide and the increase in DO to saturated levels across the water column of the GSL between July, 2012 and October, 2015 (Figure 2) demonstrate the disappearance of the DBL in response to the closure of culverts in the railroad causeway in late 2012 and late 2013. This change in geochemical regime was accompanied by an 88% decrease in MeHg, a 97% decrease in HgT, and a 18% decrease in DOC concentrations in deep waters of the GSL (Figures 2 and 3),

Using 3D analyst tools in ArcGIS (ESRI 2015), the volume of the DBL prior to causeway sealing was estimated by integration of the bathymetric surface (Baskin 2005) and the maximum elevation of the DBL surface (1272.6 meters), estimated as 2.5 meters

above the deepest point of the GSL. Multiplying this volume (1.00 km^3) by the average concentrations of HgT and MeHg in the DBL from Johnson et al. (2015) yielded approximately 54 kg HgT and 25 kg MeHg in the DBL prior to its disappearance (Table 2). Dispersing these masses into the overall water column of the South Arm (1.14e14 L, at a lake elevation of 1279 meters), the expected concentration into a homogenous water column would result in approximately 14 ng/L HgT and 7.3 ng/L MeHg, whereas the actual changes measured following the disappearance of the DBL were a roughly 8 ng/L decrease in unfiltered HgT in surface waters and a decrease in unfiltered MeHg of about 3 ng/L. Likewise, mixing the pool of DOC in the DBL into the entire water column of the GSL would be expected to increase DOC in surface waters by 6.6 mg/L, whereas the observed increase in DOC was twice the expected increase at roughly 13 mg/L.

The difference between the expected and observed increase in the concentration of unfiltered HgT in surface waters of the GSL following the disappearance of the DBL indicates that roughly 90 kg of HgT were lost from the water column to the sediments, to the atmosphere via volatilization, or via some other loss mechanism. The observed decease in HgT in both the DBL (Figure 3) and surficial sediment (Figure 4) in response to the disappearance of the DBL demonstrate that HgT was not removed from the water column by particle settling or partitioning into the sediments, suggesting that the Hg loss most likely occurred via volatilization of Hg(0) to the atmosphere, although this was not measured. Similarly, the difference between the expected increase and observed decrease in the concentration of unfiltered MeHg in surface waters of the GSL following the disappearance of the MeHg rich DBL indicates that as much as 31 kg of MeHg was lost from the water column. The decrease in the concentration of MeHg in surficial

sediments over this same time period (Figure 4) indicates this MeHg was not lost due to sedimentation or sediment partitioning, and instead was presumably lost via *in-situ* demethylation or bioadvection out of the lake. Previous research found that MeHg production in the GSL was more significant in surficial sediment relative to the DBL (Johnson et al, 2015). This result combined with our observations suggest that the DBL, when present, acts as a sort of cap that promotes the accumulation of both HgT and MeHg in both surficial sediment and the DBL. We speculate that removal of the DBL allowed for the more facile migration of MeHg and other forms of Hg from the sediment directly into surface waters, which in turn facilitates their more rapid loss from the system, such as via photo-demethylation of MeHg and volatilization of Hg(0) out of surface waters. It is unclear if the disappearance of the DBL also resulted in a decrease in net Hg methylation in surficial sediment due to the change in redox conditions and the decrease in concentrations of HgT and labile organic carbon, or if the decrease in MeHg observed in the lake sediment was purely due to the increased transport of MeHg out of the surficial sediment.

Concentrations of HgT in adult brine flies (*Ephedra spp.*) collected along the South Arm of the GSL under pre- versus post-causeway sealing conditions were lower in June following the sealing, but were higher in the other summer months after the causeway was sealed (Figure 5). Brine fly development progresses through larval and pupal stages in the lake before the adult brine flies emerge. Brine flies in all three stages of their life cycle are present at the GSL throughout the year, with larval densities lowest and pupal densities highest in June when adult emergence approaches its annual maximum (Collins, 1980). There are 1 to 2 generations of *Ephydra* per year in the GSL,

indicating that brine flies sampled during 2014 were not part of the same cohort as those sampled in 2015. On average, adult *Ephydra* survive for less than a week, and unlike other species in the *Ephydra* genus (Herbst, 1990), feeding by adult brine flies of the *Ephydra* species at the GSL is thought to be limited due to the limited availability of suitable food (Collins, 1980a, 1980b). If *Ephydra* at the GSL do not feed extensively as adults, then most Hg bioaccumulation from their diet occurs primarily during the larval stage. The extent to which the changes in HgT concentrations in the adult brine flies reflect the influence of the disappearance of the DBL versus other seasonal or interannual influences is unclear.

None of the four avian species sampled exhibited a statistically significant change in HgT when comparing samples collected before and after the disappearance of the DBL. While this may have been expected for some duck species due to a lack of connection between the locations where they forage (wetlands and bays not subject to major influence or inflows from the South Arm) and open waters of the South Arm, this was somewhat unexpected for the northern shovelers, which are believed to spend a larger portion of their foraging time in open waters of the South Arm consuming aquatic invertebrates. The diets of the four waterfowl species tend to overlap in the summer when food resources are more abundant; however, during the winter, this overlap decreases as each species either specialized on a particular resource or shifts to a previously underutilized habitat (DuBowy 1988).

Northern shovelers (*Anas clypeata*), which showed the highest HgT concentrations in this study, feed higher on the food web and move further west in the fall and winter than the other species in this study, feeding primarily on aquatic

invertebrates from open water regions of the GSL, principally brine shrimp (*Artemia*), brine shrimp cysts, brine fly larvae, and water boatman (*Corixidae)* (Vest and Conover, 2011) similar to the diet of northern shovelers reported for other areas of the US (DuBowy, 1985; Euliss et al., 1991). The other duck species sampled in our study have more generalized feeding habits and lower trophic status. Cinnamon teal (*Anas cyanoptera)* mainly feed on submerged aquatic vegetation, emergent and aquatic invertebrate larvae and pupae, and seeds in wetlands and shallower water along the lake (DuBowy 1985; Gammonley, 1995), but switch to feeding on the open water of the GSL in autumn. Gadwall (*Anas strepera)* and mallard (*Anas platyrhynchos*) had the lowest HgT concentrations and both primarily feed on freshwater plants and seeds in wetlands rather than the open-GSL, with minimal consumption of invertebrates (Gates, 1957; Gammonley and Laubhan, 2002; Rothschild et al. 2005). Given the diet of the cinnamon teal, gadwall, and mallard and the fact that most of their foraging does not occur in the open waters of the GSL, a change in the concentration of Hg in the open waters of the South Arm would not be expected to necessarily be reflected in the Hg concentration in these ducks. On the other hand, the diet and greater utilization of open water of the GSL for foraging by northern shovelers might be expected to make it more likely that any changes in MeHg concentrations or cycling in the South Arm would be reflected in the HgT levels in this avian species. However, we measured an increase in the concentrations of HgT and MeHg in surface waters due to the disappearance of the DBL (Figure 3), while there was no measurable change in the concentration of HgT in northern shovelers (Figure 6). Our observation that an increase in the concentration of MeHg and HgT in unfiltered surface waters was not accompanied by an increase in HgT in northern

shovelers may result from their diet and foraging locations being different than suggested by previous studies, or due to the effects of time of sample collection. For example, the 2014 duck harvest occurred in the Fall, approximately 9 months following causeway sealing. While we these samples with pre-closure conditions, it is possible that they may not reflect conditions prior to causeway closure. Even in the absence of the causeway sealing, significant interannual variability in avian HgT concentrations was observed (Scholl and Ball 2008; Cline 2011; Conover and Vest 2012). It is unclear if a change in Hg levels in the avian species sampled would have been measured if samples had been collected at different times, or if other avian species that feed primarily in the open waters of the South Arm had been collected, such as eared grebes or phalaropes.

Table 2. A summary of the mass balance results for HgT and MeHg in the South Arm of the Great Salt Lake pre-closure, as well as predicted concentrations of HgT and MeHg in a homogenous water column, assuming the total masses of HgT and MeHg present before closure were dispersed throughout the water column.

		[HgT] ng/L		**[MeHg] ng/L**	
		Shallow	**Deep**	**Shallow**	**Deep**
Observed Pre- Closure Concentrations	Average	10.33	54.72	1.13	25.46
	SD	6.70	14.14	1.21	4.24
Observed Post-Closure Concentrations	Average	6.26	6.86	0.54	1.00
	SD	0.84	1.80	0.40	0.47
		HgT (kg)		**MeHg (kg)**	
		Shallow	**Deep**	**Shallow**	**Deep**
Pre-closure Masses	Average	107.69	54.72	11.73	25.46
	SD	69.85	14.14	12.59	4.24
Post-closure Masses	Average	65.20	6.86	5.67	1.00
	SD	8.70	1.80	4.17	0.47
		[HgT] ng/L		**[MeHg] ng/L**	
		Shallow	**Deep**	**Shallow**	**Deep**
Predicted Concentrations	Average	14.22		3.26	
Predicted Concentrations - Observed Post-Closure Concentrations	Average	7.96	7.36	2.71	2.26

CHAPTER 5

CONCLUSION

Destratification and disappearance of the deep brine layer in the South Arm of the GSL resulted in dramatic decreases in the concentrations of HgT and MeHg in deep waters and underlying sediment in the South Arm, coinciding with a change from reduced to oxic conditions in these deep waters. These events resulted in an increase in the concentration of HgT and MeHg in surface waters, as well as the removal of 26 kg of HgT and 57 kg of MeHg from the lake, presumably due to evasion of elemental Hg(0), internal demethylation of MeHg, and possibly bioadvection out of the lake. These observations indicate that the anoxic DBL acts as an additional reservoir through which inorganic Hg and MeHg exported from the sediments must pass in order to reach oxic surface waters where it enters the food web. Thus, the presence of the DBL allows for the accumulation of high levels of HgT and MeHg in both the DBL itself and the underlying sediment, while also reducing export of Hg and from the GSL system and the degradation of MeHg. The increase in HgT and MeHg concentration measured in surface waters of the South Arm appeared to be reflected in higher HgT concentrations in brine flies in the South Arm during most of the summer. However, concentrations of HgT in four species of ducks did not change in response to the disappearance of the DBL, although this lack of a change may reflect a lack of connection between the diet and

foraging location of the particular avian species sampled and the open water of the South Arm of the GSL.

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